The Interaction of Water and Aerosols in the Marine Boundary Layer: A Study of Selected Processes Impacting Radiative Transfer and Cloudiness

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Awards #: N00014-07-1-0277
http:// http://www.atmos.washington.edu/academic/atmoschem.html

LONG-TERM GOALS

The overarching, long-term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

OBJECTIVES

For the current reporting period, our efforts have centered on completing the analysis of the combined data sets we gathered during the CARMA-IV, CARMA-III, and CARMA-II field campaigns, and an analysis of data from the Paposo, Chile site taken during the VOCALS campaign. Our objectives for these analyses have changed somewhat from those in our original proposal in light of our findings to date. We summarize them as follows.

- Using the same receptor modeling analysis for source attribution that we employed for the size-resolved aerosol hygroscopicity (Hegg et al, 2008), develop a similar source attribution for both the CCN concentration and aerosol light scattering in the marine atmosphere off the California

Report Documentation Page

Form Approved
OMB No. 0704-0188

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED
2010		**00-00-2010 to 00-00-2010**

4. TITLE AND SUBTITLE	5a. CONTRACT NUMBER
The Interaction of Water and Aerosols in the Marine Boundary Layer: A Study of Selected Processes Impacting Radiative Transfer and Cloudiness	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **University of Washington,Department of Atmospheric Sciences,Box 351640,Seattle,WA,98195-1640**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT **unclassified**	b. ABSTRACT **unclassified**	c. THIS PAGE **unclassified**	**Same as Report (SAR)**	**8**	

coast. The analysis would encompasses the three most recent CARMA studies to permit assessment of inter-annual variability in the CCN and light scattering sources.

- Perform the same sort of CCN and light scattering source attribution done in the CARMA-studies on the data taken at Paposo during VOCALS.

- Assess the importance of elemental carbon (EC) particles as CCN using data from both the CARMA studies and, if feasible, VOCALS.

- Develop a climatology of aerosol optical properties based on data from the CARMA field studies plus data from ACE-2, ACE-Asia and RED field campaigns.

APPROACH

The source attribution of CCN and aerosol light scattering requires the measurement of both the aerosol chemical composition, aerosol light scattering and the CCN activity. The first is measured via chemical analysis of filter samples by means of a variety of analytical techniques (Gao et al, 2003), the second primarily with a TSI model 3563 integrating nephelometer and the third with the DMT Inc. CNN-100 CCN spectrometer that employs the design described by Roberts and Nenes (2005). The nephelometer measured the aerosol light scattering coefficient at wavelengths of 450, 550, and 700 nm. The spectromenter measured cumulative CCN number concentrations at nominal supersaturations of 0.2, 0.3, 0.5, 0.7 and 1.0 %. The techniques described above were deployed from an airborne platform, the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter research aircraft. This platform, and its associated facility instruments, has been described in a number of publications (e.g., Wang et al, 2002; Schmid et al, 2003; Hegg et al, 2005).Because source profiles for aerosol in this area are not known, we have not employed fully deterministic source assessment (i.e., a chemical mass balance model) but rather alternative multivariate statistical techniques, specifically, the EPA PMF 3.0 and UNMIX 6.0 models. These are operational, regulatory, receptor models used precisely to deconvolute aerosol sources when good source profiles are not available. Additionally, we have tested this approach with a data set from the arctic to ensure its feasibility (Hegg et al, 2009b) and recently employed it to look at CCN source attribution during CARMA IV (Hegg et al, 2009a).

The second objective is addressed by the same approach as the first, but now applied to the VOCALS data set.

The methodology for achieving the third objective involves the use of two instrument new to our program. The first of these is the annular geometry CCN spectrometer manufactured by DMT Inc. to which we alluded above. This will yield a continuous record of the CCN concentration at five supersaturations with a time resolution of about 10 minutes. This data will be compared with the concentration of EC bearing particles measured by the SP2 instrument manufactured by DMT Inc. and recently evaluated and described by Moteki and Kondo (2007). The comparison will yield the fraction of the CCN number concentration at each supersaturation that contain EC.

The final objective, the development of a climatology of aerosol optical properties is addressed in a straightforward manner using standard statistical techniques to derive the distributions of the various optical properties over the various field campaigns for which we have data.

WORK COMPLETED

The analysis of the data from the three most recent CARMA campaigns has been partially completed, with results pertinent to our first and fourth objectives listed above in hand. Indeed, work on the first objective has essentially been completed while work on the fourth is ongoing.. The second objective, analysis of the Paposo data set from VOCALS, has also been completed. The third objective has been addressed but not yet fully completed. As mentioned in the last annual report, problems with the analysis of the SP2 data have hindered the analysis. However, much of this has been resolved and some preliminary analysis completed

RESULTS

In contrast to the analysis of the CARMA IV study alone, for which the UNMIX model was used (Hegg et al, 2009a), we employed the PMF 3.0 model for the meta analysis of the combined CARMA data bases. We did this because the UNMIX model is sensitive to the particular analytes used in the input matrix and analytes differed from year to year. Comparison of the results for UNMIX and the PMF model for the CARMA IV data set revealed consistency for all major results. Similarly, the PMF model was used on the VOCALS data set to permita more meaningful comparison with the CARMA results.

The PMF analysis for the CARMA data set, while showing significant inter-annual variability in the strength of the identified sources for both light scattering and CCN activity, also suggested broad similarity in the sources from year to year. For example, in accord with earlier results for CARMA IV alone (Hegg et al, 2009a), all years show essentially three main sources for both aerosol light scattering and CCN activity, marine, pollution and biomass burning. The year to year variability in these three sources is shown in Figure 1 for light scattering and Figure 2 for CCN activity.

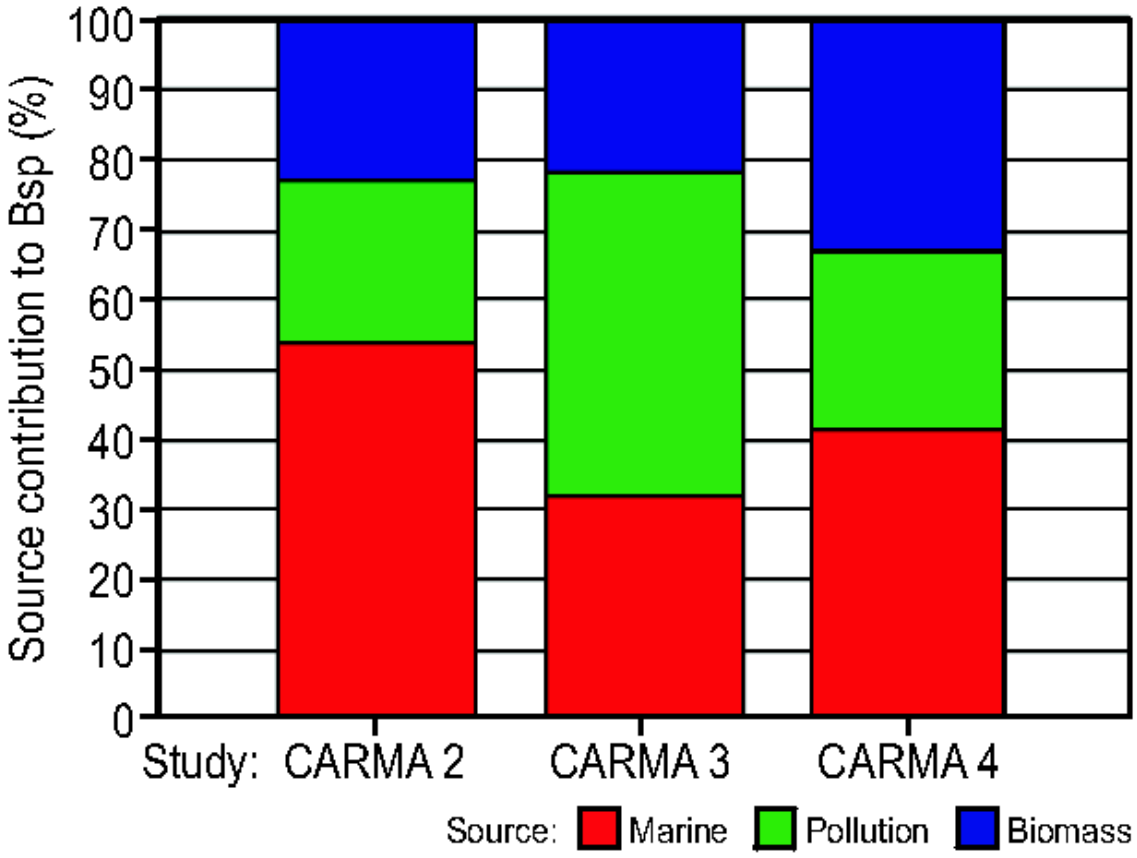

Figure 1 Bar graph showing source contributions to the aerosol light scattering for three different years during the CARMA field campaign. The marine source was the strongest during CARMA II, the pollution source the strongest during CARMA III and the marine and biomass sources the strongest during CARMA IV.

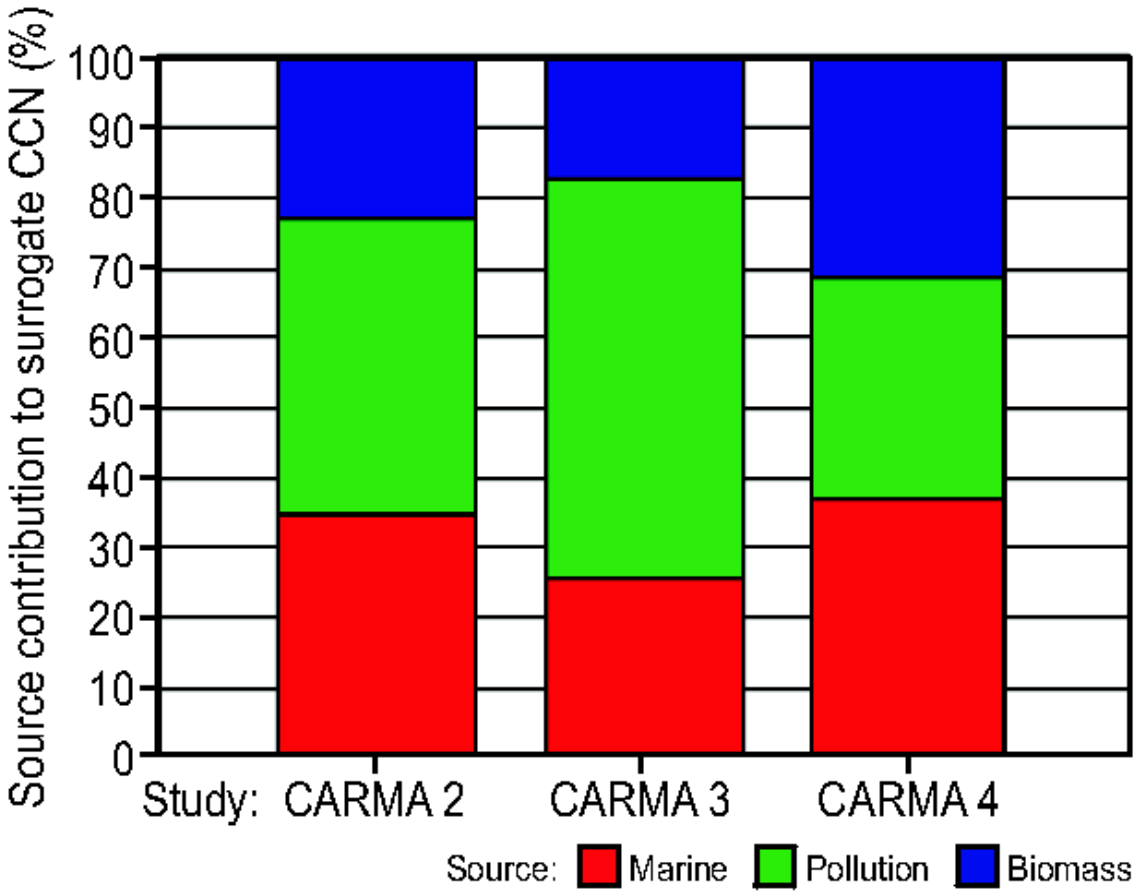

Figure 2. Bar graph showing the source contributions of the three main sources of CCN for three different years during the CARMA field campaign. For CARMA II, the largest source was pollution; for CARMA III, pollution dominated; for CARMA 4, marine aerosol was most important, closely followed by biomass burning.

For the VOCALS data set, some interesting contrasts can be seen in comparison to the CARMA results. The PMF source attribution averaged over all samples for aerosol mass, light scattering and CCN activity are shown in Figure 3. In contrast to the CARMA results, in which marine aerosol contributed ~ 40% to both light scattering and CCN activity, it only contributed around 10% to these properties at Paposo during VOCALS. The pollution sources, with perhaps a significant contribution from soil dust, dominated the aerosol properties. This arises not only from the relative strength of these sources in the VOCLAS study area but also from the meteorology of the study area, constrained as it is by the proximity of the Andes escarpment and the consequent channeling of flow on and offshore of the narrow coastal plain.

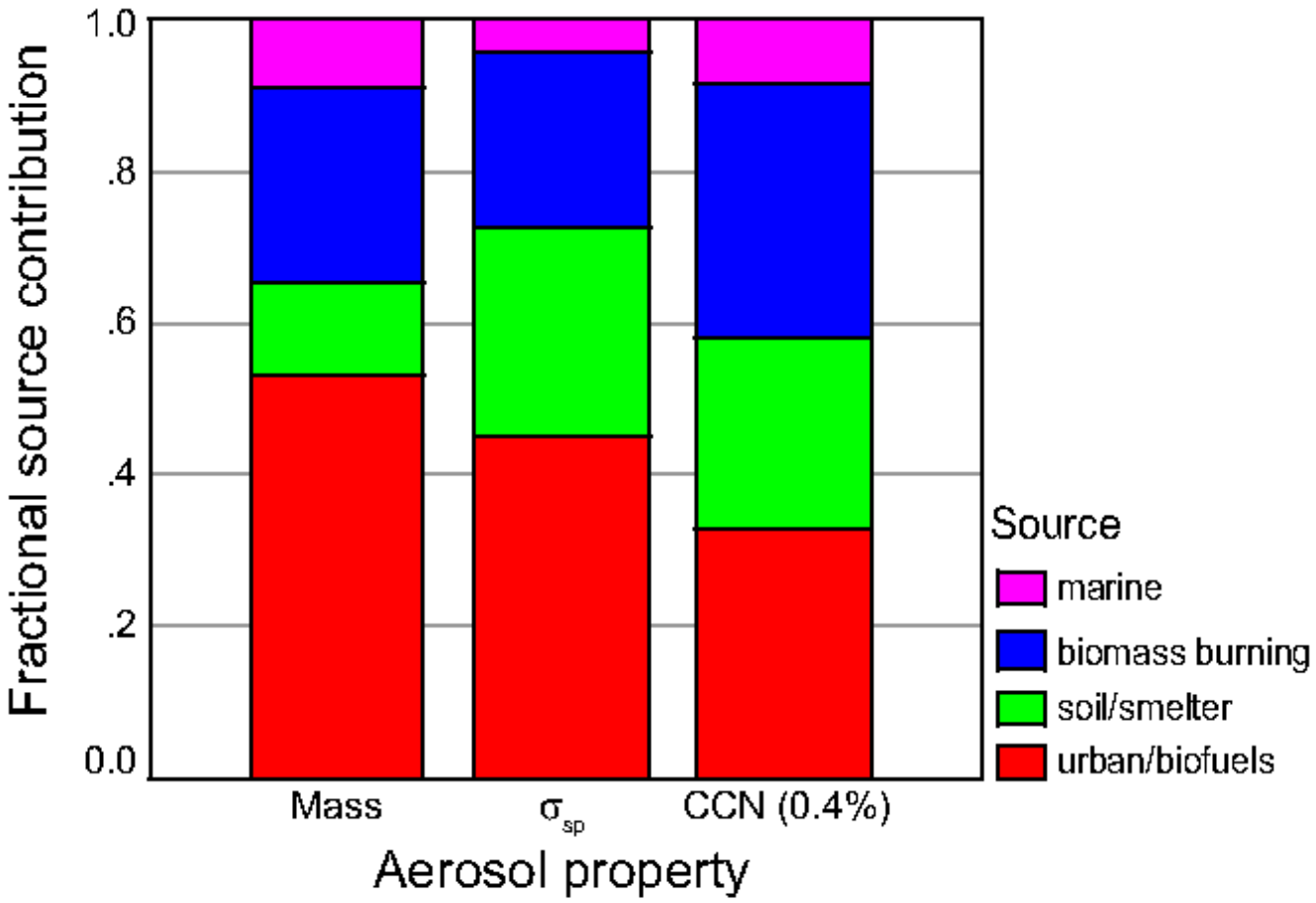

Figure 3. Bar graph showing the source contributions of the main sources of aerosol mass, light scattering and CCN activity.For aerosol mass and light scattering, the most important source was an urban pollution biofuels mix while for CCN activity this source and biomass burning were equally important.

IMPACT/APPLICATIONS

The results of the CCN and aerosol light-scattering source analysis from the CARMA studies support the conceptual picture of a mixed source aerosol in the littoral area off the California coast. The ability of the receptor model(s) to differentiate between natural and anthropogenic aerosols – and their key properties - suggests it will be a valuable tool in determining the impact of anthropogenic aerosol on both CCN activity and aerosol optical depth, the two main components of aerosol climate forcing.

TRANSITIONS

None.

RELATED PROJECTS

These measurements are highly relevant to determination of aerosol light scattering in the MBL (and thus radiative transfer in the MBL), and CCN activity (and thus of the microphysics of MBL clouds). Furthermore, numerical transport models now incorporate aerosol sources and can predict the amount of aerosol from the incorporated sources present at any particular point in the model domain. The ability to differentiate between CCN and aerosol light scattering from different sources should permit a source-specific characterization of these key aerosol properties to use in such transport models, thus improving their prognostic power.

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